

Sodium Bose-Einstein Condensates in the $F=2$ State in a Large-volume Optical Trap

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We have investigated the properties of Bose-Einstein condensates of sodium atoms in the upper hyperfine ground state in a purely optical trap. Condensates in the high-field seeking $|F=2, m_F=-2\rangle$ state were created from initially prepared $|F=1, m_F=-1\rangle$ condensates using a one-photon microwave transition at 1.77 GHz. The condensates were stored in a large-volume optical trap created by a single laser beam with an elliptical focus. We found condensates in the stretched state $|F=2, m_F=-2\rangle$ to be stable for several seconds at densities in the range of 10^{14} atoms/cm³. In addition, we studied the clock transition $|F=1, m_F=0\rangle \rightarrow |F=2, m_F=0\rangle$ in a sodium Bose-Einstein condensate and determined a density-dependent frequency shift of $(2.44 \pm 0.25) \times 10^{-12}$ Hz cm³.

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So far, Bose-Einstein condensation in dilute atomic gases [1, 2, 3, 4, 5] has been achieved in all stable bosonic alkali isotopes except ³⁹K and ¹³³Cs, as well as in atomic hydrogen [6] and metastable helium [7, 8]. The physics that can be explored with Bose-Einstein condensates (BEC) is to a large extent governed by the details of interatomic interactions. At ultra-low temperatures, these interactions not only vary significantly from one atomic species to another but can also change significantly for different internal states of a single species. While in ⁸⁷Rb, only minor differences of the collisional properties are observed within the ground state manifolds, in ⁷Li, the magnitude of the scattering length differs by a factor of five between the upper and the lower hyperfine manifold and even the sign is inverted [9]. The behavior of ²³Na with a scattering length of 2.80 nm in the $|F=1, m_F=\pm 1\rangle$ states and 3.31 nm in the $|F=2, m_F=\pm 2\rangle$ states [10] is intermediate between these two extreme cases. Thus, sodium might provide a system in which the study of BEC mixtures of states with significantly differing scattering length is possible. Such a mixture would be a natural extension of earlier work on spinor condensates in ⁸⁷Rb [11, 12] and in the $F=1$ manifold of ²³Na [13, 14].

In this Letter, we report the realization of Bose-Einstein condensates of ²³Na in the upper $F=2$ hyperfine manifold in a large-volume optical trap [15]. In ⁸⁷Rb, condensates in both the $F=1$ and $F=2$ states had been achieved by loading atoms in either state into a magnetic trap and subsequent evaporative cooling. In contrast, sodium BECs have previously only been produced in the $F=1$ state. Early attempts at MIT and NIST to evaporatively cool sodium in the $F=2$ state were discontinued since the evaporative cooling scheme proved to be more robust for the $F=1$ state. Instead of developing an optimized evaporation strategy for $F=2$ atoms in a magnetic trap, we took advantage of an optical trap which traps atoms in arbitrary spin states [16]. After producing $F=1$ condensates and loading them into an optical trap, we

transferred the population into the $F=2$ manifold using a single-photon microwave transition at 1.77 GHz. We found that a BEC in the stretched $|F=2, m_F=-2\rangle$ state is stable on timescales of seconds at densities of a few 10^{14} atoms/cm³. Simultaneous trapping of condensates in the $|2, -2\rangle$ and $|1, -1\rangle$ states for several seconds was also achieved. In contrast, at the same density, a condensate in the $|2, 0\rangle$ state decays within milliseconds. Nevertheless, we were able to observe the so-called clock transition $|1, 0\rangle \rightarrow |2, 0\rangle$ in a BEC, which is to lowest order insensitive to stray magnetic fields. By taking spectra of this transition at various condensate densities, we were able to measure a density-dependent frequency shift of $(2.44 \pm 0.25) \times 10^{-12}$ Hz cm³.

The basic setup of our experiment is described in [17, 18] and is briefly summarized here. We have prepared condensates of more than 4×10^7 ²³Na atoms in a so-called ‘clover-leaf’ magnetic trap with trapping frequencies of $\nu_x = 16$ Hz and $\nu_y = \nu_z = 160$ Hz by radiofrequency evaporation for 20 s. After preparation of the condensate in the $|1, -1\rangle$ state, the radial trapping frequencies were adiabatically lowered by a factor of 5 to decompress the condensate. Subsequently, an optical trapping potential was superimposed on the condensate by slowly ramping up the light intensity. After turning off the remaining magnetic fields, nearly all atoms were loaded into the large-volume optical dipole trap. The resulting peak density reached 5×10^{14} atoms/cm³, slightly higher than the density in the magnetic trap.

The large-volume optical trap was realized by shaping the output of a Nd:YAG laser (typically 500 mW at 1064 nm) with cylindrical lenses leading to an elliptical focus with an aspect ratio of approximately 25. At the location of the condensate, the focal size was $\approx 20 \mu\text{m}$ along the tight axis resulting in an optical trapping potential with typical trap frequencies of $\nu_x = 13$ Hz axially and $\nu_y = 36$ Hz and $\nu_z = 850$ Hz transversely. The trap axis with the largest trapping frequency was oriented verti-

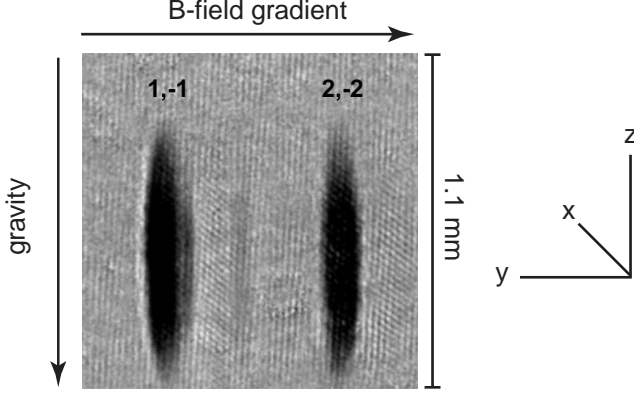


FIG. 1: Sodium condensates in the $|1,-1\rangle$ and $|2,-2\rangle$ state, 30 ms after release from the trap. After preparation of the mixture the atoms were held in the optical trap for 1 s. The horizontal separation of the spin states is due to application of a magnetic-field gradient during expansion.

cally to counteract gravity. The pancake shape of the trap, which we had recently used to create (quasi-) 2D condensates [18], provided a much larger trapping volume than our previous cigar-shaped optical traps [16, 19] and thus significantly larger condensates could be stored.

Optically trapped condensates were observed by absorption imaging on the closed $|F=2, m_F=-2\rangle \rightarrow |F'=3, m_F'=-3\rangle$ cycling transition at 589 nm after sudden release from the trap, using light propagating parallel to the trap laser. The ballistic expansion time was typically 30 ms, after which the vertical size of the condensate had increased by more than a factor of 100 while the horizontal expansion was less than a factor of two. To make sure that atoms in both the $F=1$ and the $F=2$ manifold could be detected simultaneously, a short laser pulse resonant with the $F=1 \rightarrow F'=2$ transition was applied to pump all atoms into the $F=2$ manifold. State-selective detection could be achieved by applying a magnetic field gradient of several G/cm during the free expansion of the atomic cloud, leading to a spatial separation of spin states which differ in the orientation of the magnetic moment (Fig. 1).

In order to test the intrinsic stability of the optical trap, we first investigated the lifetime of condensates in the $|1,-1\rangle$ state as shown in Fig. 2 a). Even after 70 s of dwell time, more than 10^6 atoms remained in the condensate. Generally, the decay of the number of atoms N in the condensate can be modelled by the rate equation

$$\frac{dN}{dt} = -k_1 N - k_2 N \langle n \rangle - k_3 N \langle n^2 \rangle, \quad (1)$$

where k_1, k_2, k_3 are the one-, two- and three-body loss coefficients and n is the condensate density. By setting either k_2 or k_3 to zero in Eq. 1, analytical fitting functions for the decay of the condensate number can be derived. A fit to the data in Fig. 2 a) with $k_2 = 0$ and a one-body loss rate $k_1 = 0.029 \text{ s}^{-1}$ (determined by an exponential fit

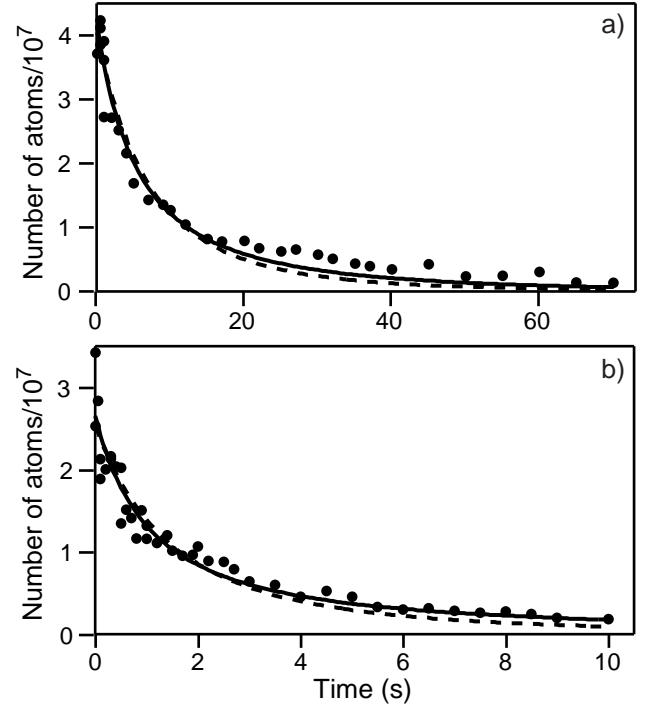


FIG. 2: Lifetime measurement of sodium BECs in the $|1,-1\rangle$ (a) and $|2,-2\rangle$ (b) states in a large-volume optical trap with an initial peak density of $\approx 5 \times 10^{14} \text{ atoms/cm}^3$. The number of atoms is determined from the measured size of the clouds after ballistic expansion. The lines are fits where either only three-body loss (solid) or two-body loss (dashed) has been assumed in addition to a one-body loss rate of 0.029 s^{-1} .

to all data points with $t \geq 40 \text{ s}$) yields a three-body loss coefficient of $k_3 = (1.57 \pm 0.17 \pm 0.55) \times 10^{-30} \text{ cm}^6 \text{ s}^{-1}$, in agreement with the previously published value [16]. Here, the first error is the statistical error of the fit and the second error represents systematic uncertainties originating from the measurements of the cloud sizes and the determination of k_1 . Since the densities in the present work are lower than in previous work, two-body loss processes might also contribute. A fit where k_3 was set to zero yielded a two-body rate coefficient of $k_2 = (3.98 \pm 0.47 \pm 0.81) \times 10^{-16} \text{ cm}^3 \text{ s}^{-1}$, larger than theoretical predictions [20]. The fits indicate that the observed loss is predominantly due to three-body collisions, though two-body processes cannot be ruled out.

Condensates in the $|2,-2\rangle$ state were produced by applying a microwave pulse at 1.77 GHz to an optically trapped $|1,-1\rangle$ condensate. By varying power and duration of the microwave pulse, we were able to adjust the ratio of atoms transferred into the $|2,-2\rangle$ state between 0 and 100 %. Fig. 2 b) shows a measurement of the lifetime after complete transfer into the $|2,-2\rangle$ state. The lifetime in the $|2,-2\rangle$ state is still on the order of seconds but it is significantly shorter than the lifetime of a $|1,-1\rangle$ condensate (Fig. 2 a). The reduced lifetime can be attributed to much larger three- and/or two-body loss

rates. Using the solutions of Eq. 1 we deduce rate coefficients for the atom loss, assuming that only one process is responsible for the loss. Thus, we obtain as upper bounds $k_2 = (2.93 \pm 0.28 \pm 0.29) \times 10^{-15} \text{ cm}^3 \text{ s}^{-1}$ and $k_3 = (1.53 \pm 0.13 \pm 0.32) \times 10^{-29} \text{ cm}^6 \text{ s}^{-1}$. Both values are in reasonable agreement with theoretical predictions [21, 22]. Though, at typical densities, the decay rate in the $F=2$ state is roughly an order of magnitude larger than in the $F=1$ state, it should still be compatible with direct condensation in the $F=2$ manifold, provided that the loss coefficients for the magnetically trapable $|2,+2\rangle$ state are similar to those for the $|2,-2\rangle$ state.

By transferring only part of the atoms into the upper hyperfine manifold we could also observe mixtures of condensates in the $|1,-1\rangle$ and $|2,-2\rangle$ states (see Fig. 1). In the presence of small magnetic field gradients, we observed a rapid spatial separation of the two components in a time shorter than 100ms due to the fact that the $|1,-1\rangle$ state is low-field seeking while the $|2,-2\rangle$ state is high-field seeking. During the separation, strong density modulations in both components were observed, which could be attributed to tunnelling processes playing a role in the separation process [23]. Afterwards, the two components lived almost independently side by side in the trap and the individual lifetimes were not significantly affected. When we tried to compensate all stray magnetic field gradients, we still found that in steady state the two components tend to separate, i.e. we observed domains with only one component [14]. This indicates that the two states are intrinsically not miscible. While we found ^{23}Na BECs in the $|2,-2\rangle$ state as well as mixtures of $|1,-1\rangle$ and $|2,-2\rangle$ condensates to be stable for several seconds, non-stretched states in the $F=2$ manifold as well as $F=1$, $F=2$ mixtures with $|m_1 + m_2| \neq 3$ decayed within several ms for typical condensate densities on the order of $10^{14} \text{ atoms/cm}^3$. This fast decay is probably due to (two-body) spin-relaxation which is strongly suppressed in ^{87}Rb but occurs with rate constants on the order of $10^{-11} \text{ cm}^3 \text{ s}^{-1}$ in ^{23}Na [21].

A particularly interesting transition within the electronic ground state of alkali atoms is the magnetic-field insensitive transition $|F,0\rangle \rightarrow |F+1,0\rangle$, often referred to as clock transition since its equivalent in cesium is used as the primary time standard. Shortly after laser cooling had been realized, the benefits of using ultracold atoms for atomic clocks had become apparent [24] and today the most accurate atomic clocks are operated with laser-cooled atoms [25]. Therefore, it seems natural to investigate the use of a BEC with its significantly reduced kinetic energy for the study of the clock transition.

To observe the clock transition, we first completely transferred an optically trapped $|1,-1\rangle$ condensate into the $|1,0\rangle$ state with a radiofrequency Landau-Zener sweep. Selective driving of the $|1,-1\rangle \rightarrow |1,0\rangle$ transition was achieved by applying a 3 G offset field which provided a large enough quadratic Zeeman-shift to lift

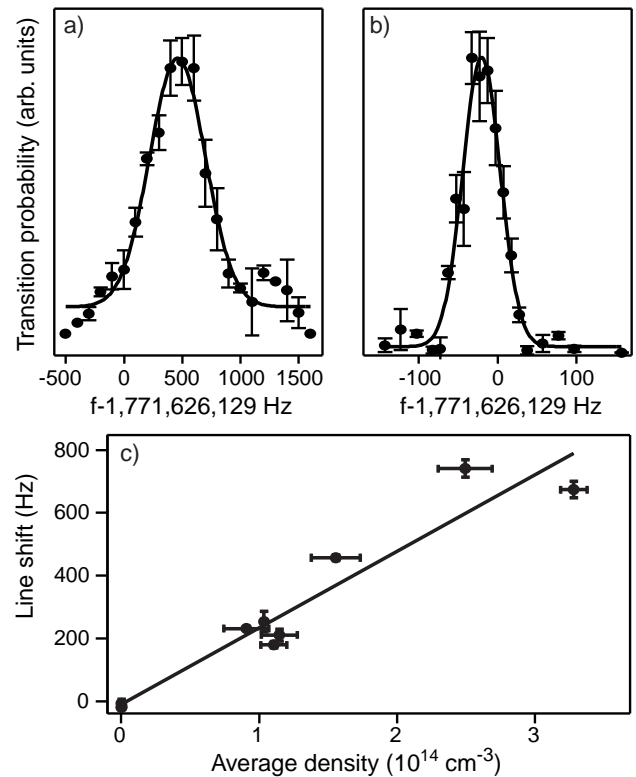


FIG. 3: Magnetic-field insensitive transition $|1,0\rangle \rightarrow |2,0\rangle$ in a BEC. (a) Spectrum in the trap at a mean density of $1.6 \times 10^{14} \text{ atoms/cm}^3$. (b) Spectrum after 12.5 ms time-of-flight at a mean density of $4.3 \times 10^{11} \text{ atoms/cm}^3$. The discrepancy between the center of the line and $\nu = 0$ is probably due to an error in the exact determination of the residual magnetic field. The solid lines are Gaussian fits. (c) Transition frequency as a function of density yielding a clock shift of $(2.44 \pm 0.25) \times 10^{-12} \text{ Hz cm}^3$.

the degeneracy with the $|1,0\rangle \rightarrow |1,+1\rangle$ transition. Subsequently, the magnetic field was reduced to a value of typically 100 mG which keeps the spins aligned and gives rise to a quadratic Zeeman shift of the clock transition of $\approx 20 \text{ Hz}$. The $|1,0\rangle \rightarrow |2,0\rangle$ transition was then excited by using a microwave pulse at 1.77 GHz with a duration between 2 and 5 ms. The fraction of atoms transferred into the $|2,0\rangle$ state was kept below 20% in order to ensure a practically constant density in the $|1,0\rangle$ state during the pulse. Immediately afterwards, the optical trap was turned off suddenly and the number of atoms which made the transition was detected by state-selective absorption imaging after 15-30 ms of ballistic expansion. A typical spectrum showing the number of transferred atoms as a function of microwave frequency (corrected for the calculated quadratic Zeeman shift) for a BEC with an average density of $1.6 \times 10^{14} \text{ atoms/cm}^3$ is shown in Fig. 3a). The density was determined by measuring the release energy [18] of $|1,-1\rangle$ condensates without applying a microwave pulse. The release energy E_{rel} is related to the chemical potential

μ by $E_{rel} = (2/7)\mu = (2/7)(h^2 a_{|1,-1\rangle|1,-1\rangle} / \pi m) n_0$ [26]. Here, $a_{|a\rangle|b\rangle}$ is the scattering length between two ^{23}Na atoms in states $|a\rangle$ and $|b\rangle$ ($a_{|1,-1\rangle|1,-1\rangle} = 2.80\text{ nm}$), m is the ^{23}Na mass, h is Planck's constant and n_0 is the peak density in the condensate related to the average density by $\bar{n} = (4/7)n_0$. The spectrum in Fig. 3a) is significantly broadened compared to the one in Fig. 3b), which is taken after ballistic expansion, and the transition frequency is shifted with respect to the unperturbed frequency $\nu_0 = 1,771,626,129\text{ Hz}$ [24].

In the limit of weak excitation, the density-dependent shift of the clock-transition frequency is due to the difference in mean-field potential that atoms in the $|1,0\rangle$ and $|2,0\rangle$ state experience within a $|1,0\rangle$ condensate. Taking into account the inhomogeneous density distribution of a trapped BEC, this leads to a line shape given by [27]

$$I(\nu) = \frac{15h(\nu - \nu_0)}{4n_0\Delta U} \sqrt{1 - \frac{h(\nu - \nu_0)}{n_0\Delta U}} \quad (2)$$

with

$$\Delta U = \frac{h^2}{\pi m} (a_{|2,0\rangle|1,0\rangle} - a_{|1,0\rangle|1,0\rangle}), \quad (3)$$

where the center of the line is at $\nu_0 + 2n_0\Delta U/3h$ and the average frequency is $\nu_0 + 4n_0\Delta U/7h$. In our experiment, the line is additionally broadened and the asymmetry of Eq. 2 smeared out due to the finite width of the microwave pulse which was limited by rapid inelastic losses in the $|2,0\rangle$ state. Therefore, we have used a (symmetric) Gaussian to fit the resonances where we have identified the fitted center frequency as the average frequency of the line. By taking spectra of the clock-transition at different densities we have determined a density shift of $(2.44 \pm 0.25) \times 10^{-12}\text{ Hz cm}^3$ (Fig. 3c). Here, the error is the statistical error from a linear fit to the data. Additional systematic errors due to fitting of the line with a Gaussian and due to an uncertainty in the determination of the density are estimated to be smaller than 20%. Using Eq. 3 and $a_{|1,0\rangle|1,0\rangle} = 2.71\text{ nm}$ [10], we determine the scattering length $a_{|2,0\rangle|1,0\rangle} = 3.15 \pm 0.05\text{ nm}$ for collisions between two atoms in states $|1,0\rangle$ and $|2,0\rangle$.

In conclusion, we have prepared condensates in the upper $F=2$ hyperfine manifold of the sodium ground state in a large-volume optical trap and observed a stable condensate in the high-field seeking stretched state $|2,-2\rangle$. Since only the stretched state exhibits reasonable stability, experiments with more complex spinor condensates do not seem to be possible. Furthermore, we have for the first time observed the alkali clock-transition in a Bose-Einstein condensate and determined the value for the density-dependent mean-field shift. In present BEC

experiments, the magnitude of the shift precludes the use of trapped condensates for precise atomic clocks. However, under circumstances where the condensate density can be drastically reduced as may be feasible in space-based experiments, the extremely low velocity spread of BECs might help improve the accuracy of atomic clocks.

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